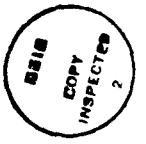
STUDIES OF COMPOUND STATES OF ATOMIC NEGATIVE IONS USING LASER BEAMS*

Office of Naval Research Task NR 393-071



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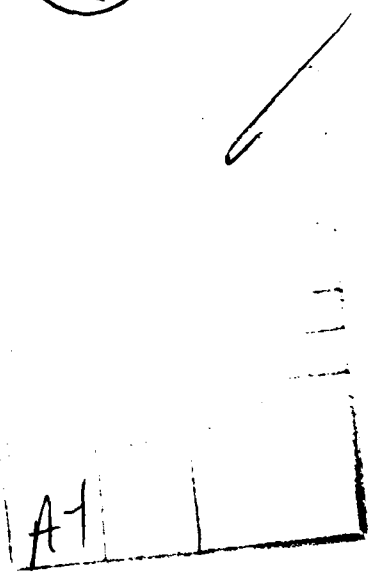

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Prepared in cooperation with Dr. D. J. Pegg and Mr. J. Thompson (University of Tennessee) and with Mr. M. J. Shea (Graduate Student, Vanderbilt University).

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Laser photodetachment of negative ion beams. Neutralization of negative ion beams. Autodetachment spectroscopy of negative ion beams. Electron collisions in strong laser fields.

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

This research involves experimental studies of compound states of atomic negative ions using laser beams. Negative ion beams of stable (H^-, O^-, etc.) and metastable (He^-, Be^-, Ca^-, etc.) species are generated by double charge exchange from positive ion beams colliding with alkali vapor. Measurements of the photodetachment and autodetachment spectra for these ions provide new information on compound state energy levels. In addition, new experiments are underway to

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study the collisions of electrons with atoms in strong laser fields. Many theoretical predictions have been presented recently on this subject. These experiments should provide the first tests of such predictions.

STUDIES OF COMPOUND STATES OF ATOMIC NEGATIVE IONS USING LASER BEAMS

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This is a first-year progress report on our research effort entitled "Studies of Compound States of Atomic Negative Ions Using Laser Beams" under the Office of Naval Research (ONR) task NR393-071. Our proposal and research is neatly divided into two related efforts — (I) Autodetachment and Laser Studies of Compound States of Be^- and (II) Electron Collisions in the Field of a Laser. During the past year considerable experimental progress has been made on both of these projects. Prior to receiving ONR funding, R. N. Compton (Health and Safety Research Division) and G. D. Alton (Physics Division) Oak Ridge National Laboratory (ORNL), and D. J. Pegg of the University of Tennessee Physics Department have collaborated without direct funding to perform lifetime, photo-, auto-, and collisional detachment spectroscopy measurements of He^- ions using the ion source of the EN tandem accelerator. However, due to tandem accelerator commitments, this necessitated research one week at a time every few months. Thus, the experimental setup had to be assembled and disassembled during each scheduled week of research — an inefficient means of doing research. After one year of experimental setup, we have modified the existing negative ion source test facility for use in negative ion atomic physics research. This facility is available for use for extended periods of time. The negative ion test facility has been funded principally by the Physics Division. The ONR support has provided some operating funds and, most importantly, the funds for Dr. T. J. Kvale, a postdoctoral fellow from Professor J. T. Park's laboratory at the University of Missouri. Dr. Kvale joined our group January 1, 1984, and is now primarily responsible for the day-to-day operation of the negative ion test facility, when used for atomic physics research. In addition, we have received some in-house seed support for this project which has allowed us to independently develop a high-intensity, high-resolution laser-based pulsed electron source. A graduate student from Vanderbilt University, Michael Shea, has been hired to assist in carrying out these studies. Mr. Shea began work in December 1983 after spending three months as a Great Lakes Colleges

Association/Associated Colleges of the Midwest (GLCA/ACM) student on the project before the seed money was received. Mr. Shea will return to Vanderbilt University in September to begin his graduate studies. At that time a third-year graduate student from Yale University, Mr. Perry Blazewicz, will begin a one-year project on this work.

We will divide the discussion of the research into two parts. The first part (I) summarizes the negative ion test facility studies and constitutes the main part of the work. The second part (II) describes the progress in developing the pulsed electron source and the construction of a trochoidal electron spectrometer.

I. Studies of Negative Ion States

We have made major modifications to the negative ion source test facility in order to perform the proposed Be^- experiments. The negative ion source test facility, shown schematically in Fig. 1, is a multiple-purpose facility with emphasis placed on extending our knowledge of methods and mechanisms for negative ion generation and of the structure and properties of negative ions.

Figure 1 illustrates the basic components which make up the facility. The facility can be operated for the generation and acceleration of positive ions which can be double charge-exchanged to produce negative ions prior to or following momentum analysis. In the former case, positive ions produced in a hollow cathode positive source are accelerated to energies up to 30 keV for singly charged particles focused through a recirculating Li charge-exchange cell where they are converted to negative ions through double charge exchange. Negative ions, thus created, are postaccelerated by the accelerator tube assembly and injected into the analyzing magnet at energies up to 150 keV. Postmomentum analysis charge-exchange can also be effected by reversing the polarities of the ion source housing and magnet and focusing the analyzed positive ion beam through the Li charge exchange cell situated immediately before the experimental chamber. In this way, positive ion beam energies up to 150 keV for singly charged particles can be obtained. The latter technique will permit observation of very short-lived negative ion states in our experimental apparatus. The facility can also be equipped with direct extraction type negative ion sources for producing an extensive number of high-intensity negative ion beams.

Extensive modifications have been made to the facility in order to perform a variety of atomic physics experiments involving negative ions. Specifically, we have fabricated, installed, and tested a universal charge-exchange negative ion source which is presently being used to generate positive and negative ions. The charge-exchange source consists of a positive ion hollow cathode source, ion extraction electrode system, einzel lens, and recirculating type Li charge exchange cell. Negative ions can be formed in either of two cells located prior to or following magnetic analysis. The post-analysis charge-exchange cell is located in close proximity to the experimental chamber so that we can study metastably bound negative ions with lifetimes as short as a few tens of nanoseconds. This arrangement should enable us to search for very short-lived autodetaching negative ion states. In addition to the previously described modifications to the facility, we have converted the magnetic mass analyses system so that circularly symmetric beams can be formed at the plane of analysis. (This modification was necessary in order to optimally transport the ion beam to our experimental chamber.) We have, as well, fabricated and installed beam transport components lenses, magnetic steerers, valves, drift tube, and aperture, postanalysis charge-exchange cell and an experimental chamber which houses specially designed electrostatic deflection system, Faraday cups, and electrostatic energy analyzer for performing the intended research.

A schematic drawing of the experimental apparatus is shown in Fig. 2. This arrangement permits generation and study of short-lived negative ion states as previously mentioned. Thus, the negative, positive, and neutral components are separated by the computationally designed deflection system. The negative and positive beams are monitored in shielded and biased Faraday cups. A portion of the negative ion beam is allowed to pass through a small aperture located in the center of the first Faraday cup and enters a gas cell which is used to collimate the beam and produce collisionally detached electrons. The ion beam passes through a spherical sector electrostatic analyzer and is monitored in the rear Faraday cup. Detached electrons accompanying the ion beam are energy analyzed with the spherical sector electrostatic energy analyzer in the fixed pass energy mode. In this mode of operation electrons moving slower than the pass energy of the spectrometer are accelerated, while those moving faster than this pass energy are decelerated. In this way, our spectrometer resolution function remains constant.

In addition to the rather extensive changes in the negative ion source test facility, we have converted our data acquisition system from analog to digital. The data acquisition system, shown schematically in Fig. 3, centers around a LeCroy 3500c CAMAC based data acquisition and control microcomputer and/or multichannel analyzer system. The system has available 64 K of RAM and 30 K of PROM firmware for data acquisition, manipulation, and experimental apparatus control. The data acquisition time for accumulation in a particular channel are determined by monitoring the transmitted ion beam in the second Faraday cup and digitizing the analog signal which is fed into a preset scaler. After accumulation of a predetermined amount of charge, a signal from the preset scaler is used to inhibit further data acquisition, store, and to advance a multichannel scaling unit to the next channel where the process is repeated. Signals derived from the CAMAC based multichannel scaling unit are used to advance a locally constructed external DAC which drives a bipolar operational amplifier used to change the voltage on the electrostatic energy analyzer. The electron signal, which is transmitted through the spectrometer, is detected with a conventional channel electron multiplier/amplifier circuit and fed into a CAMAC based ADC where it is stored in the memory of the multichannel analyzer.

During the past year we have published in *Physical Review* part of our earlier He^- studies. This work is attached as Appendix I. A second paper on collision detachment of H^- and He^- is finished and has been submitted to *Physical Review A*. We will not go into the details of these experiments here.

The He^- studies are almost complete. Only two rather minor studies remain. (1) As we reported in the *Physical Review* article (Appendix I), we have observed significant blackbody photodetachment of He^-. We have built a liquid-N_2 beam shroud which will help to quantify these studies. This should be a relatively easy experiment. (2) Since the photodetachment cross section of He^- is large at long wavelength, it should be possible to measure the photodetachment threshold for He^- by passing a tunable infrared light source (dispersed filament blackbody radiation). Bunge and Bunge (private communication) predict that the EA[He(1s2s)] = 77.53 ± 0.04 (meV). Thus, we should expect a photodetachment threshold at 159,890 Å. Since the photodetached electrons are collected along the beam direction, this allows us great efficiency in

detecting the slow electrons which will result from the photodetachment. Also, the infrared light beam can be sent down the length of the He^- ion beam to increase the interaction length. The liquid nitrogen shroud should reduce the background blackbody radiation photodetachment.

The primary aim of the presently proposed work is, however, to produce and study Be^- ions. The negative ion states of Be have been of considerable interest both theoretically and experimentally. Single particle scattering resonances for group-II elements (Mg, Zn, Cd, and Hg) have been measured by Burrow et al.[1] Unfortunately, they were unable to study the simplest and theoretically most tractable system, namely Be, due to the difficulty in generating Be beams and its health related hazards. Negative Be ions were first detected in a mass spectrometer by Bethge et al.[2] and it is now possible to make microampere beams of Be^- by double charge-exchange of Be^+ ions.[3,4] Thus, the only experimental information on negative ions of Be is that long-lived Be^- exists. Theoretically, the situation regarding Be^- is confusing. Various methods have been employed to calculate the energy and width of the $Be^-(1s^22s^22p)$ shape resonance. Hunt and Moisewitsch[5] use a simple model potential to predict $E_r = 0.6$ eV and $\Gamma = 0.22$ eV. Rescigno et al.[6] use the complex-coordinate method to obtain $E_r = 0.76$ eV and $\Gamma = 1.11$ eV and Kurtz and Ohrn[7] use the so-called Harris method employing an ad hoc polarization potential from which they obtain $E_r = 0.195$ eV and $\Gamma = 0.283$ eV using static exchange polarization. Semiempirical extrapolation methods give values for E_r between 0.19 and 0.68 eV.[8-12] One of the most surprising predictions of the extrapolation techniques is the prediction that $Be^-(1s^22s^23s)$ lies below $Be^-(1s^22s^22p)$ and, in fact, is bound! (See Refs. 8, 10, 11, and 12.) This prediction is rather startling although Bates[13] pointed out in 1947 that, in special cases, an s electron may be more tightly bound to a neutral atom than a p electron because of the short range nature of the potential field and the greater penetratibility of an s orbital. Quoting Massey[14] "... for Be^-, the ground state is $3s^2S_{1/2}$ and is almost certainly stable whereas the $2p^2P^0_{1/2}$ is unstable." Thus, it is possible that the Be^- ions observed in the mass spectrometer experiments are $Be^-(1s^22s^23s)$. Unfortunately, the *ab initio* calculations of Jordan and Simons[15] find no evidence for a stable $(1s^22s^23s)^2S$ state of Be^-. Another possibility for stable Be^- ions is a long-lived Feshbach resonance like the $^4P^o$ state of He^-. A likely candidate is $Be^-(1s^22s2p^2)^4P^e$ (see Ref. 15).

We are now in a position to use the Negative Ion Source Test Facility and the apparatus described above to determine, by direct measurement, whether Be^- is metastable. We can measure a lifetime in the range from 10^{-8} s to 10^{-4} s. Provided that Be^- is metastable we will determine the autodetachment electron energy as was done recently for He^- (see Appendix I). If the ion is $Be^-(1s^2 2s 2p^2)$, as suggested by Weiss,[16] the autodetachment electron energy will be ~2.5 eV. Our experiment would give the energy level of Be^- to within ±0.1 eV.

On the basis of our studies so far, these experiments should be straightforward. We have gained considerable expertise in this area over the past three years. The experimental details are given in our earlier publications[17-19] which are attached to this proposal. We should emphasize our unique capability for determining autodetachment electron energies for metastable negative ions. After one year of experimental setup and preliminary testing of the apparatus, we are now in a position to complete this study.

In addition, we propose to photodetach electrons from Be^- and determine the photoelectron energy distribution with the photoelectron spectrometer used in the autodetachment experiments. These data taken together with the autodetachment energies will provide an exact energy level for Be^-. The photoelectron energy will be calibrated using photoelectrons from O^-. The O^- reference beam can be easily introduced into the apparatus at any time. We will also measure the photodetachment cross section for Be^- in the energy range from 0.6 to ~7.8 eV. Other shapes and Feshbach resonances which are coupled to the metastable Be^- state and lie in the range of the tunable dye laser can also be studied. In addition, it should be possible to observe photodetachment of $Be^-(1s^2 2s 2p^2)$ into both $Be(1s 2s^2 2p)^3P^o$ and $Be(1s^2 2p^2)^3P^e$ final states using the output of the (F_2) excimer laser at 1570 Å(7.89 eV). The two photoelectron energy peaks will be separated in energy by exactly 4.675 eV. The observation of these two peaks would provide unambiguous identification of the state of Be^-.

II. Electron Collisions in the Laser Field

In our original proposal we described a set of experiments involving laser-induced transitions between compound negative ion states. Two basic experiments were proposed: (1) laser-induced

transitions between the He^- ($1s2s^2$) and higher doublet states, and (2) resonant laser-induced negative ion production.

We have made considerable progress in performing these two tasks. We have constructed and put into operation two new apparatus for performing these experiments. The first involves a trochoidal electron spectrometer capable of merging an electron beam, alkali or gas (He) beam, and a laser beam over a path length of ~10 cm. The second experiment involves the development of a novel new laser produced pulsed electron source. We will discuss each of these experiments below.

A schematic of the trochoidal electron spectrometer is shown in Fig. 3. The apparatus has been successfully operating for about one month. The $He^-(1s2s^2)$ ion resonance is clearly observed and is used to calibrate the electron energy scale. A graduate student from Vanderbilt University, Mr. Larry Hudson, is presently working on the apparatus with Dr. J. A. Stockdale. This project may result in his Ph.D. thesis. This apparatus will be used to study two of our proposed experiments. In the first experiment, the electron beam will be tuned to the $He^-(1s2s^2)$ resonance at 19.34 eV and a laser will be tuned to higher doublet He^- ion states. Slow electrons will be detected as "trapped electrons" when the laser excites an upper state which decays by leaving an excited He atom and a slow electron. This experiment is now in operation and should provide some results within the next year.

One of the major problems in studying electron collisions in the field of a laser is the fact that most high peak power lasers are pulsed with typical durations of $\sim 10^{-9}$ s. Such experiments also require large electron fluxes during the time that the laser is on. In this connection we have developed a novel pulsed source of high intensity monoenergetic electrons using a pulsed laser. Although this work is a part of the present proposal, it was necessary to obtain seed money support from ORNL to complete these studies. Mr. Mike Shea, a graduate student from Vanderbilt University, has been working on this project.

In several branches of pure and applied science there is a critical need for a high-brilliance source of monoenergetic well-collimated electrons. Many of these applications also require pulsed electron beams with high repetition rates and short pulse times. Perhaps one of the most noteworthy examples of this need is in the study of collision phenomena in the field of a laser.

During the past five years there have been many theoretical calculations concerning the ability of high-power lasers to alter chemical reactions. The most elementary of these reactions is the subject of this proposal, electron chemistry in a laser field. Since most high-powered lasers are pulsed beam devices, it is desirable to have a source of pulsed electrons in experiments which involve simultaneous laser and electron beams. A continuous thermionic electron source of, say, 10^{-3} amp could only produce on the order of 10^{6} electrons during a typical laser pulse (~10 ns). A pulsed electron source of the same average current could deliver 10^{12} electrons during the laser pulse.

The traditional approach to the production of monochromatic electron beams has been the selection of a narrow slice of electrons from a thermionic cathode. The best reported energy width of an electron beam produced by an electrostatic energy analyzer is ~3.5 meV, whereas the best reported resolution obtained in an actual scattering experiment is ~20 meV.

A photoionization source for the production of monoenergetic electrons is superior to the thermionic source for a number of reasons. First, the photoionization source produces electrons which have a very low initial energy spread which can *eliminate* the need for a monochromator. This feature is especially significant if the metal atoms that form the photocathode are in the gas phase as opposed to a metallic surface. This is an important point since the monochromator can introduce energy and angular aberrations in an electron beam. Secondly, the space charge limited current for a 180° electrostatic energy analyzer is proportional to the energy spread to the five-halves power whereas for the photoionization source the current is directly proportional to the energy spread, ΔE. This means that the monochromator current varies as $(\Delta E)^{5/2}$ whereas the photoionization current depends only linearly upon ΔE. Therefore, the photoionization source is expected to be superior for energy widths below ~0.05 eV.

Electrostatic electron spectrometers never produce the theoretical energy resolution for which they are designed. One major reason for this failure to realize optimum resolution is the fact that the electron beam is no longer in thermodynamic equilibrium after acceleration or deceleration. Coulomb interactions between the electrons result in an energy broadening which is proportional to the beam density and the initial energy spread. This broadening occurs within the analyzer as well

as in the drift paths. Electron focusing through slits also increases the energy spread. It is, therefore, desirable to keep the distance between source and target as short as possible. The photoionization source should be ideally suited for this purpose.

Gallagher and York[20] have constructed a photoionization electron spectrometer in which a cw laser was made to photoionize a metastable ($^1D_2$) barium beam. The beam intensity was 10^{-12} to 10^{-13} amp and an ultimate resolution of ~1 meV was obtained under optimum conditions. This apparatus was quite complicated and involved the use of a low-powered He-Cd laser to produce the 325-nm photoionization laser beam. The technique described in this proposal has a number of advantages in simplicity of design and ease of operation over that of the metastable barium photoionization method. One major difference, of course, is the pulsed nature of our proposed source. Finally, a French group is presently developing a pulsed photoionization electron beam using the light from a synchrotron radiation source. Average beam currents are only $\sim 10^{-14}$ amp and are not likely to increase much above 10^{-12} amp. We do not look upon this technology as a serious competition to our technique.

We have received, free gratis, a Lumonics excimer laser from the Oak Ridge Gaseous Diffusion Plant. This laser was a fallout of the now defunct Molecular Laser Isotope Separation Group. After some initial effort, the laser has been functioning daily for approximately eight months. The initial experiments involved the construction of an apparatus consisting of an atomic beam of Cs passing between two parallel plates with a hole in one plate through which electrons can be accelerated. The electron beam was characterized with respect to angular divergence, pulse width, and energy spread. The energy spread was approximately determined by passing the electrons through a cell and noting the steep rise in electron current with energy. The derivative of this function gives an upper limit to the electron resolution. By studying the transmitted electron current through the cell with He or Ar gas present as a function of electron energy, resonances will be detected at the position of the He^- or Ar^- states. This should allow for considerable accuracy in determining the absolute energy and energy resolution of the electron beam.

We have made very large electron currents by using the 308 nm XeCl radiation crossing the Cs beam. The excimer laser power is ~50 mJ/pulse corresponding to $\sim 10^{17}$ photons/pulse. The

photoionization cross section for Cs is 2×10^{-19} cm^2 at $\lambda = 308$ nm. Since the product of cross section and laser fluence is of the order of unity, most of the Cs atoms in the laser beam are ionized by the laser pulse. Thus, in the absence of recombination we should expect between 3×10^{11} electrons/pulse/cm^3 to $\sim 2 \times 10^{14}$ electrons/pulse/cm^3, depending upon the sample temperature. The measured electron currents are about two orders of magnitude less than this corresponding to instantaneous currents of $\sim 10^{-3}$ amp. We have further found that surface ionization by a laser pulse yield considerably higher electron yields (tens of amperes), but with an energy spread far greater than that available from a properly designed metal vapor source. There may be many applications including the present work where large electron fluxes are needed. Electron ejection from a surface by 308 nm light would certainly be a convenient and efficient such source of electrons.

A patent application has been initiated on this novel new electron source.

During the next year we propose to use these two new electron scattering apparatus for the purpose of studying electron chemistry in the field of a laser. In particular, we will investigate the laser excitation of $He^-(1s2s^2)$ in the trochoidal spectrometer as discussed above. The new pulsed electron source will be used to make Na^- transient negative ions from which we will study the radiative stabilization of Na, via the process $e + Na \rightarrow Na^{*-} \rightarrow Na^- + 2h\nu$. Calculations for such a process have been performed by M. H. Mittleman.[21]

Budget

A. Support for Personnel (Including Fringe Benefits and Overhead)

	Second Year	Third Year
R. N. Compton (5%)	$10,000	$12,000
G. D. Alton (5%)	10,000	12,000
Postdoctoral Fellow (100%)	41,000	41,000
B. Ancillary Cost		
Technical Support	6,000	6,000
Travel	1,000	1,000
Channeltron Detector	1,000	1,000
Publication and Supplies	1,000	1,000
TOTAL	$70,000	$74,000

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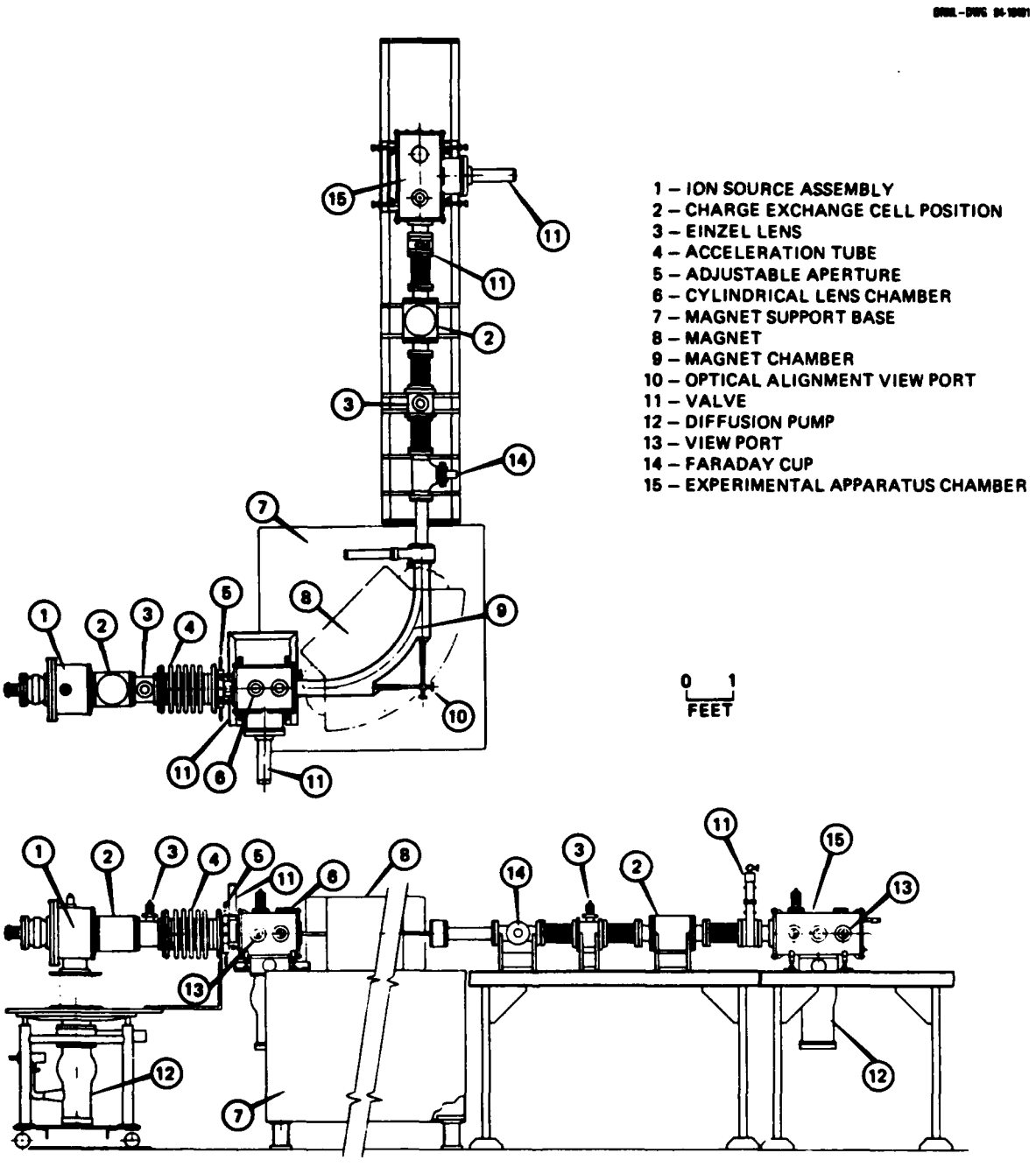

Fig. 1. Schematic of the Negative-Ion-Source Test Facility.

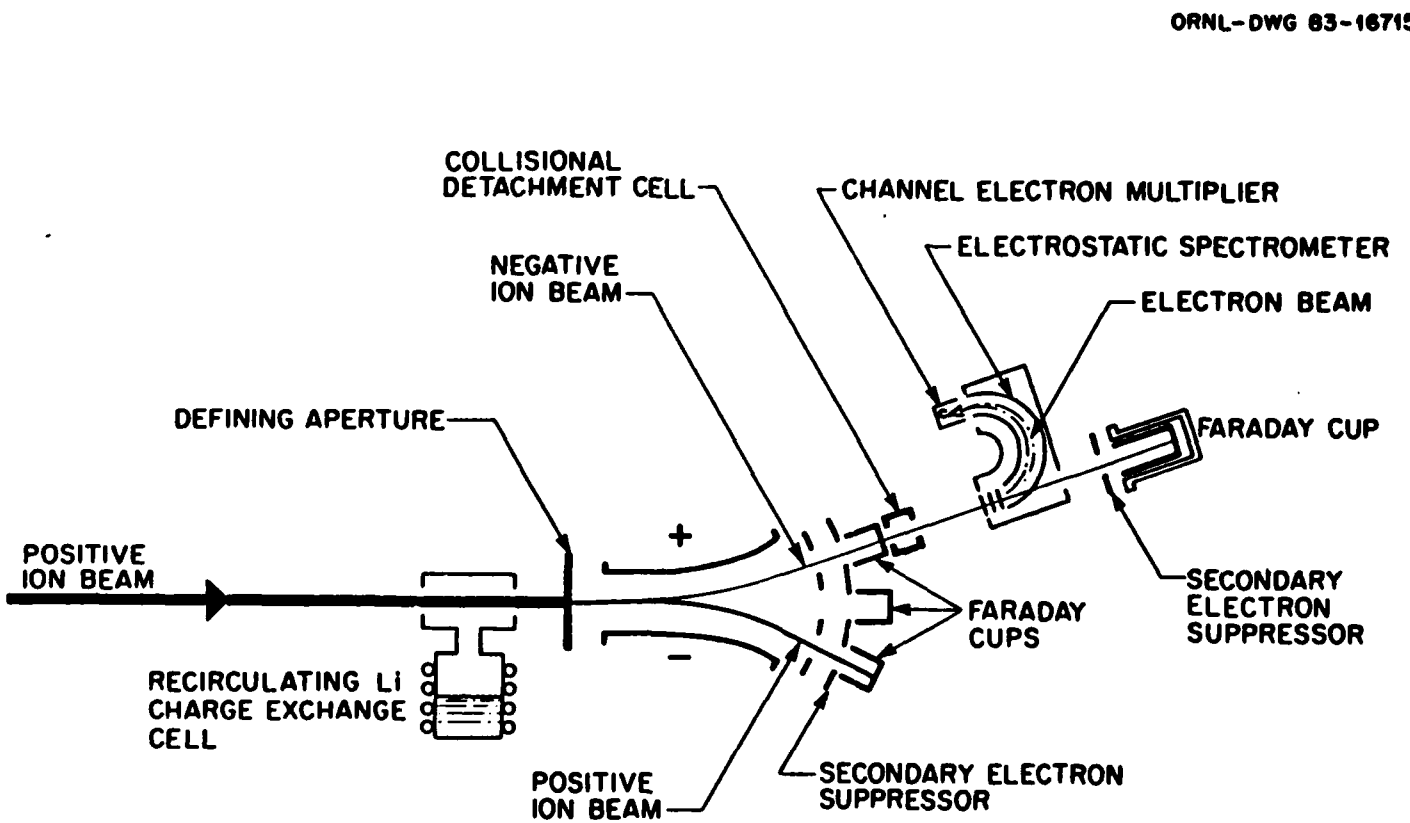

Fig. 2. Experimental Apparatus for Measuring Short Half Life Metastable Ions.

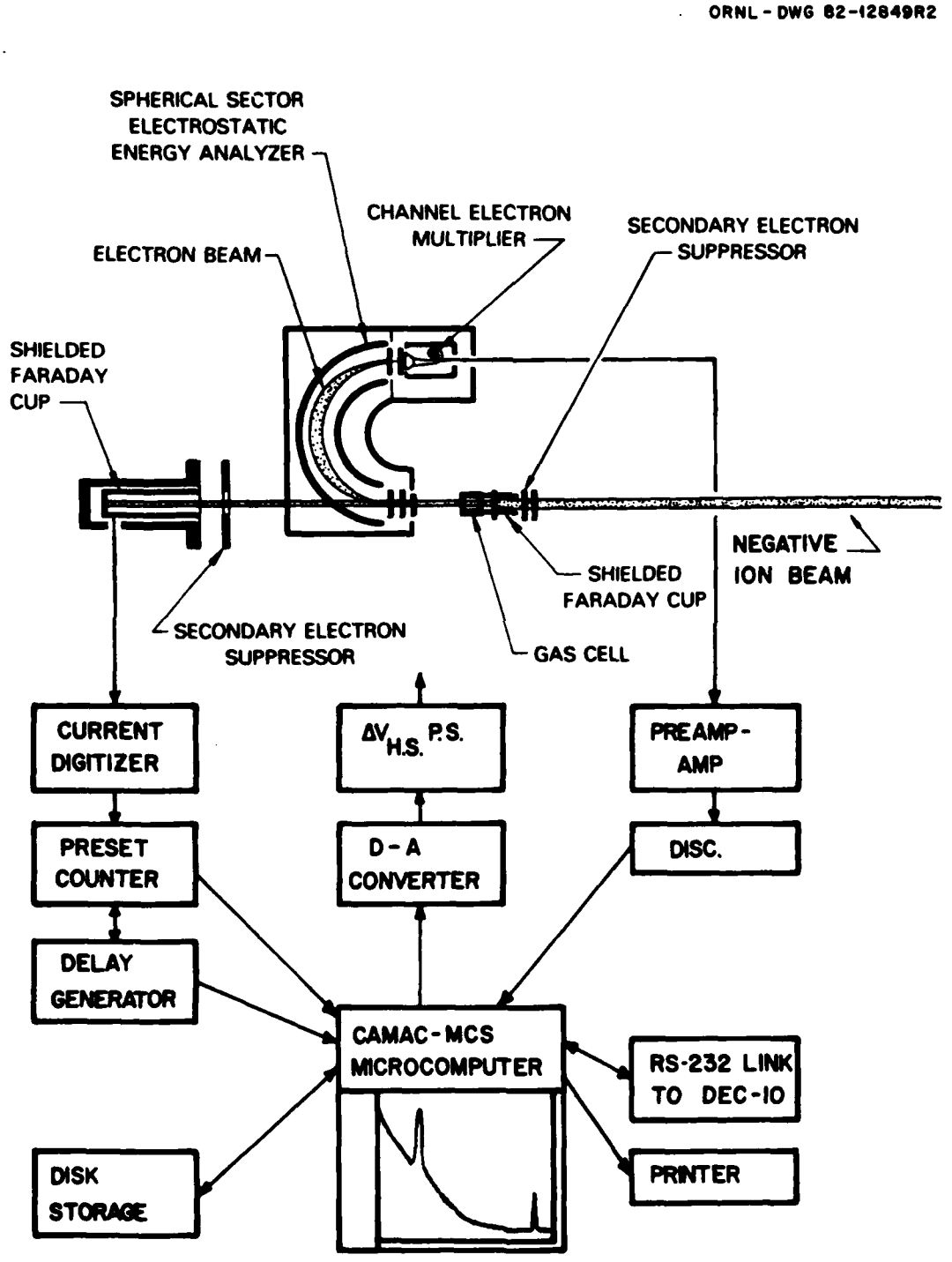

Fig. 3. Data Acquisition System.

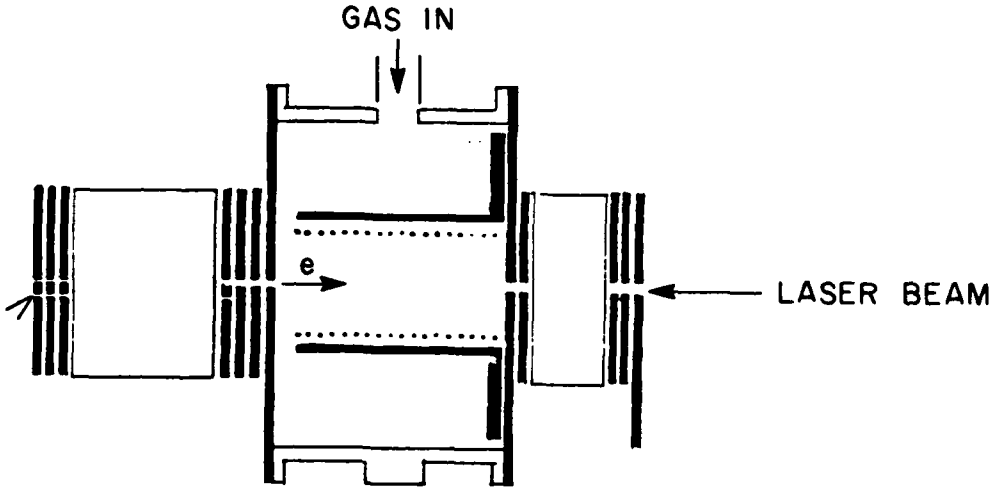

Fig. 4. Trochoidal Electron Beam-Laser Beam Apparatus. The Electron Beam and Laser Beam are Collinear.

RESUME

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Photography

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Concerning a long-lived $(^2P^e)He^-$ state: Lifetime and energy-level measurements of He^-

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The question concerning the existence of a long-lived doublet $(^2P^e)He^-$ state has been addressed by measuring the lifetime and energies of autodetaching electrons from 20–120-keV He^--ion beams formed by charge exchange in Ca vapor—a vapor conducive with $(^2P^e)He^-$ formation. The measured lifetime and electron energy levels agree closely with accepted values for components of the $(^4P^o_J)He^-$ states. No other structures were detected in the electron energy spectrum encompassing the anticipated energy level of the autodetached electron from the $(^2P^e)He^-$ state.

I. INTRODUCTION

The negative–helium-ion state is a classical example of a core excited state which is bound within the detachment continuum of the neutral atom. It has, therefore, been the subject of many investigations directed toward elucidating its configuration identification,[1] lifetime against autodetachment,[2–5] energy levels,[1,6–11] fine-structure separations,[12,13] and destruction characteristics by electric field,[9,10,14] collisional,[15,16] and photoelectron-detachment processes.[17,18] Because of the nature of the forces involved in this weakly bound atomic system, it has served as a basis for development and testing of sophisticated wave functions and atomic Hamiltonians necessary for a proper description of the observed states of the system.

Theoretical studies of He^-, predating experimental detection of the ion, were made by Wu who used variational-type wave functions in investigating the stability of the $(1s2s^2)^2S$ He^- electronic configuration.[19] Evidence of a long-lived He^- state was first presented by Hiby who, while analyzing the negative-ion mass spectrum extracted from a helium plasma discharge, detected a weak signal in the mass-4 position.[20] This discovery subsequently gave impetus to a number of experimental and theoretical studies of the He^- ion.

The first plausible prediction of a He^- state with long-lived properties was made by Holøien and Midtdal who used multiterm variational-type wave functions to study the binding properties of various electronic configurations of the ion.[1] The $(1s2s2p)^4P^o_J$ He^- metastable states were found to be bound relative to the $(1s2s)^3S$ He atomic configuration by 0.075 eV and stable against direct Coulomb autodetachment as well as electric-dipole radiative-decay processes. The long lifetime characteristics of the state were explained according to the *LS*-coupling scheme by virtue of the fact that the respective $J=\frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$ components could only decay through weak spin-dependent interactions. Furthermore, the respective fine-structure components were postulated to have significantly different lifetimes because of the possibility of coupling between the $^4P_{1/2,2/3}$ and $^2P_{1/2,3/2}$ states of the same configuration—the latter of which are allowed to decay promptly via the Coulomb interaction. Thus, the lifetimes of these states would be decreased relative to the pure $J=\frac{5}{2}$ component.

The theoretical calculations of Manson[12] indicate that the energies of the fine-structure levels of the $(^4P^o_J)He^-$ state increase with increasing J and have energy excesses of the $\frac{5}{2}$ level above the respective $\frac{3}{2}$ and $\frac{1}{2}$ levels of 8.43×10^{-6} and 3.89×10^{-5} eV. The experimental measurements of Mader and Novick[13] using magnetic-resonance techniques yielded values of 3.46×10^{-6} and 3.58×10^{-5} eV for the same energy excesses—which are in reasonable agreement with the theoretical estimates.

The advent of fast and larger computer facilities has enabled the use of more sophisticated wave functions for more accurate theoretical estimates of the binding energies and fine-structure separations of the $(^4P^o)He^-$-ion states. Table I summarizes the results of several theoretical estimates of the binding energy for the $(^4P^o)He^-$ states. The degree of progress in terms of accuracy is exemplified by the recent configuration-interaction calculations of Bunge and Bunge[8] in which a value of 0.077 eV was obtained for the binding energy of the $(^4P^o)He^-$ relative to the $(^3S)He$ state. The results agree closely with the experimental value of 0.076 eV obtained by Oparin *et al.*[10] using electric field detachment techniques and the 0.080-eV value determined by Brehm, Gusinow, and Hall[11] using laser photoelectron-detachment techniques.

The discovery and development of an efficient forma-

TABLE I. Summary of theoretically determined energy levels for $^4P^o_J$ states of He^-.

$E[(1s2s)^3S]-E[(1s2s2p)^4P]$ (eV)	$E[(1s2s2p)^4P]$ (eV)	Reference
≥ 0.075	19.744	1 (Theor.)
≥ 0.033	19.786	6 (Theor.)
0.067	19.752	7 (Theor.)
0.0774	19.7417	8 (Theor.)

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tion technique for producing relatively intense ion beams in the long-lived He^- state through charge-exchange interactions between keV He^+ ions and gaseous or vaporous materials has greatly aided experimental investigation of the properties of the ion as well as enabled its use in high-energy–accelerator applications.[21,22] The technique is especially efficient whenever low–ionization-potential exchange vapors are utilized. The dominant process for $(^4P_J^o)He^-$ formation with Group-IA exchange vapors, as demonstrated experimentally,[21] is through sequential exchange collisions between energetic He ions and atoms of the exchange medium. The process proceeds according to the following spin-conserving reactions:

$$(1s\,^2S)He^+ + (^2S)X \rightarrow (1s2s\,^3S)He + (^1S)X^+ ,$$

$$(1s\,2s\,^3S)He + (^2S)X \rightarrow (1s2s2p\,^4P^o)He^- + (^1S)X^+ ,$$

where X represents the exchange atom. The latter interaction occurs with high probability due to the close proximity of the potential-energy curves for the entrance $[(^3S)He]$ and exit $[(^4P)He^-]$ channels.

According to theoretical prediction, the $J=\frac{1}{2}$ and $\frac{3}{2}$ levels of the $(^4P^o)He^-$ state are expected to decay at significantly faster rates than that of the $J=\frac{5}{2}$ level due to their coupling to the corresponding $(^2P^o)He^-$ levels of the same configuration.[1] The necessity of including coupling to such states considerably complicates theoretical calculations of the lifetimes for the $J=\frac{1}{2}$ and $\frac{3}{2}$ components. On the other hand, there are no states of the same configuration with which the $J=\frac{5}{2}$ level can couple and, therefore, the interaction Hamiltonian is explicitly known—making estimates of the lifetime of this metastable state considerably less difficult. Several theoretical estimates of the lifetime of the $J=\frac{5}{2}$ level have been made.[12,23–25] Of such calculations, the 455-μsec value determined by Labahn and Estberg[25] is considered the most accurate and is in reasonable agreement with experiment.

The admixture of several states with differential metastabilities also complicates experimental measurement of the lifetimes of the respective components. The difficulties of such determinations are exemplified by attempts to measure the lifetimes of the respective components of the $(^4P_J^o)He^-$ state.[3,5] The results of several measurements of the bound metastable states of He^- formed in a variety of exchange media are summarized in Table II.

In the presence of a uniform magnetic field, substates with the same M_J, the quantum number of the component of J which lies along the magnetic field direction, can be mixed (Zeeman effect). This effect will be more pronounced for states which lie close to each other and, hence, mixing between the $J=\frac{1}{2}$ and $\frac{3}{2}$ levels of the $(^4P_J)He^-$ states can occur at relatively low magnetic field strengths resulting in a decrease in population of the $J=\frac{5}{2}$ level and an increase in that of the $J=\frac{1}{2}$ level. Under such conditions, the average lifetime of the He^- ion will be decreased. Of the values shown in Table II, magnetic-momentum analyses were utilized in the experiments of Nicholas *et al.*[2] and Simpson *et al.*,[4] and, thus, one might expect an increase in the population of the lower-lying levels at the expense of the $J=\frac{5}{2}$ level in these experiments. This effect will be significant even at low magnetic field strengths.[5] The magnetic field mixing effect was utilized by Blau, Novick, and Weinflash,[3] and Novick and Weinflash[5] to separate the various fine-structure states, thus permitting estimations of their individual lifetimes.

TABLE II. Summary of experimental results of previous measurements of the lifetimes of metastably bound $^4P_{1/2,3/2,5/2}$ states of He^-.

Source of He^-	Lifetime (μsec)	Reference
$He^+ + He$	18.2	2
$He^+ + K$	11.5 ± 5, 345 ± 90	3
$He^+ + N_2$	9^{+5}_{-3}	4
$He^+ + K$	10 ± 2, 16 ± 4, 500 ± 200	5

In spite of the large number of investigations that have taken place concerning the long-lived He^- state, questions still exist regarding the exact identity of the observed state or states. More specifically, plausible arguments have been made by three experimental groups[26–28] that a long-lived $(1s\,2p^2)^2P^e$ He^- may exist in addition to the well-established $(1s\,2s\,2p)^4P^o$ He^- state which was generally assumed to be the only long-lived negative-ion state of helium. The state, if bound, would be metastable against Coulomb autodetachment and direct electric-dipole radiative-decay processes.

The first of such reports was made by Baragiola and Salvattelli[26] who used a two-state approximation in analyzing He^--formation cross-section data obtained by interacting a 20–40-keV He beam with what was believed to be a known $(1^1S,2^3S)$ mixture with an atomic Mg target. The authors concluded that the cross sections obtained from ground-state He capture were consistent with the existence of a long-lived $(^2P^e)He^-$ state. Similar conclusions were also drawn by Pedersen *et al.*[27] who applied beam-attenuation techniques to measure metastable $(^3S)He$ fractions produced by both electric field and collisional detachment of He^-. The constant metastable fraction obtained from both experiments led the investigators to conclude that the ion beam was made up of both $(^2P^e)He^-$ ($\sim29\%$) and $(^4P^o)He^-$ states ($\sim71\%$). The formation of He^- ions by interactions between fast-moving He atoms in the (^1S) and (^3S) states and H_2 target has been investigated by Dunn *et al.*[28] who show that the cross section for He^- resulting from $(^1S)He$ electron capture is approximately one-half that of capture to the (^3S) state. In this experiment, the $(^1S)He$ state was prepared by collisions between doubly charged He^{2+} projectiles and H_2 exchange-gas molecules according to the following spin-conserving reaction:

$$He^{2+} + H_2 \rightarrow (^1S)He + (^2S)H^+ + (^2S)H^+ .$$

Negative He^- ions were then produced by allowing the $(^1S)He$ to again react with H_2. The process is assumed to proceed according to the reaction

$$(^1S)He + H_2 \rightarrow [(1s\,2p^2)^2P^e]He^- + H_2^+ .$$

Thus, if total electron spin is conserved, the latter reaction leads to a doublet rather than a quartet He^- state. How-

ever, in subsequent experiments, Pedersen finds that apparent metasable (^3S)He fractions derived by use of the beam-attenuation technique do not necessarily coincide with the true values—regardless of the attenuating gas used.[29] Thus, previously reached conclusions which were based on the use of the attenuating gas H_2, must be reevaluated.

According to the first-order perturbation calculations of Safronova and Senashenka,[30] the $(^2P^e)He^-$ state is bound with an eigenenergy of 19.91 eV above the ground state of He and can decay by forbidden autodetachment processes with lifetimes of the $J=\frac{1}{2}$ and $\frac{3}{2}$ levels of 92 and 5.5 μsec, respectively, and by radiative processes with a lifetime of 0.2 μsec. Thus, the state, although bound, would effectively have a short half-life. In addition, the state, if bound, is expected to lie above the $(1s2s2p)^2P^o$ He^- state to which it can decay radiatively. The latter state has been observed in energy-dependent differential electron scattering experiments.[31,32]

The $(^2P^o)He^-$ state appears as a resonance at $\sim$0.5 eV above the threshold for excitation of the $(1s2s)^3S$ level of He at an electron energy of 20.5 eV.[31] The energy-level value of this doublet state is in exact agreement with recent theoretical calculations of Chung[33] who finds the resonance to be of Feshbach character and the state to be bound to the $(1s2s)^1S$ He state by 83 and 124 meV for the $J=\frac{1}{2}$ and $\frac{3}{2}$ levels, respectively, which decay by autodetachment of a 20.495-eV electron. The $(1s2s2p)^2P^o$ He^- state is of particular relevance to the present study in that the conjectured $(1s2p^2)^2P^e$ He^- state can radiate to this level which, in turn, can decay promptly through Coulomb autodetachment. The 20.5-eV eigenenergy of the autoejected electrons associated with the decay of the subsequently formed $(^2P^o)He^-$ state would then serve as a signature of the existence of the $(^2P^e)He^-$ state.

In the present article, we address the question of the existence of a long-lived doublet state of He^- by measuring the lifetime and electron energy spectra from a He^--ion beam produced by charge exchange in calcium vapor—a vapor which can produce both $(^2P^e)He^-$ and $(^4P^o)He^-$ states. The $(^2P^e)He^-$-formation process proceeds according to the following spin-conserving reactions:

$$(^2S)He^+ + (^1S)Ca \rightarrow (^2P^e)He^- + (^1S)Ca^{2+} ,$$

in single collisions, while $(^4P^o)He^-$ formation proceeds according to the sequential collisional processes of

$$(^2S)He^+ + (^1S)Ca \rightarrow (^3S)He + (^2S)Ca^+ ,$$

$$(^3S)He + (^1S)Ca \rightarrow (^4P^o)He^- + (^2S)Ca^+ .$$

Thus, based on these spin-conserving reactions, the doublet-state $(^2P^e)$ population should increase linearly with exchange vapor pressure p, while the quartet-state $(^4P^o)$ population should increase as p^2. If we relax the Wigner spin-selection rule, $(^4P^o)He^-$ can be formed also in the former reaction and the $(^2P^e)He^-$ can be formed in the latter reaction as well.

II. LIFETIME MEASUREMENTS

A. Experimental techniques

A negative-ion beam composed of bound metastable particles will spontaneously autodetach during free flight by electron emission and in the absence of a perfect vacuum lose additional electrons through collisions with the residual background gas. A beam of particles with initial intensity $I^-(0)$ will decrease exponentially to value $I^-(t)$ after having drifted for a time t and distance x according to the following relationship:

$$I^-(t)=I^-(0)\sum_{j=1}^{s} f_j \exp\left[-\frac{t}{\tau_j}\right]\exp\left[-\sum_{i=0}^{z}\sigma_{-1,i}nx\right],$$

where f_j is the fraction of the beam in state j with lifetime τ_j, s is the number of metastable components in the ion beam, z is the atomic number of the negative ion, and $\sigma_{-1,i}$ is the cross section for electron loss to the ith state of the projectile due to collisions with the residual gas atoms of density n. For simplicity the assumption is made that the residual gas is composed of one species only.

A schematic diagram of the apparatus utilized in measuring the average lifetime of the decaying states of He^- is shown in Fig. 1. A positive-He^+-ion beam formed in and extracted from a duoplasmatron ion source was focused through a charge-exchange cell containing chemically pure-grade Ca vapor. During all measurements, the charge-exchange cell was operated at intermediate power levels which were commensurate with the production of both $(^2P^e)He^-$ and $(^4P^o)He^-$. The power on the cell was typically maintained at values such that the total negative-He^--ion current was $\sim$10% of those normally achieved using Ca exchange vapor under equilibrium capture and loss conditions. (Total momentum-analyzed He^--negative-ion currents of 0.2 μA were typically produced compared to $\sim$2 μA for equilibrium production and loss conditions.) The region of He^- production was situated $\sim$2.5 m from the experimental measurement apparatus.

The negative-He^--ion beam emerging from the cell was increased to the desired energy by a postacceleration stage, momentum analyzed, collimated, and finally passed into the experimental apparatus. The technique employed in measuring the lifetimes of autodetaching states of He^- differs from those previously reported in that it is differential in nature rather than integral. The device utilized consisted of a 26-cm-long parallel-plate charge-collection system composed of a grounded top plate and a biasable bottom plate separated by 2.86 cm. Autodetached and collisionally produced electrons were collected on the positively biased bottom plate of the system. Negatively biased apertures were placed at the entrance and exit ends of the parallel-plate arrangement to ensure that only those electrons emitted or collisionally produced during transit across the plates were collected. The ion beam was monitored in a biased and carefully shielded Faraday cup which could be externally positioned with respect to the ion beam to compensate for beam deflection whenever voltage was applied to the bottom plate. All ion and elec-

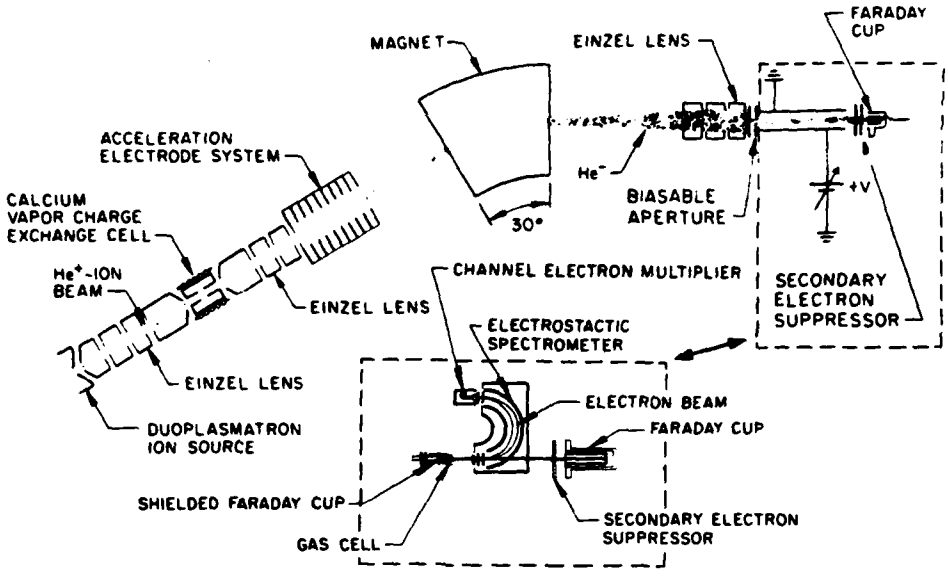

FIG. 1. Experimental arrangement for measuring the lifetimes and energy levels of metastably bound He^- formed in charge-exchange collisions with Ca vapor.

tron currents were measured with electrometers with stated accuracies of $\pm 5\%$.

In order to distinguish between autodetachment and collisional-detachment processes, both of which were present at the base pressures typically achieved ($\sim 2\times 10^{-7}$ Torr), electron-detachment data were accumulated as a function of N_2 chamber pressure for a given ion energy and a logarithmic plot of the corrected collection data versus pressure extrapolated to zero pressure. Such measurements were repeated for a range of ion energies lying between 20 and 120 keV. Corrections to the collected electron signal due to projectile ionization of the N_2 target gas were made by alternately reversing the plate polarity and measuring the collisionally produced positive-ion current. The total electron and ion currents collected during the polarity reversal cycle were then subtracted. Since the He^--ion current was measured on the exit side of the collection plate, corrections were made by adding the net electron current to the measured value.

The magnetic field flux densities required for momentum analysis of the He^--ion beams utilized in the experimental investigations ranged from 900 G at 20 keV to 2200 G at 120 keV. Thus, Zeeman mixing would be expected for closely lying levels such as those associated with the $^4P_J^o$ He^- states.

B. Results and discussions

Least-squares fits to the electron-signal collection data versus nitrogen density in the experimental chamber are shown in Fig. 2 for a number of ion energies. The consequent ordinate intercepts of these data are shown in Fig. 3 plotted against ion-beam transit time. The slope of a least-squares fit to the latter data yields the average life-

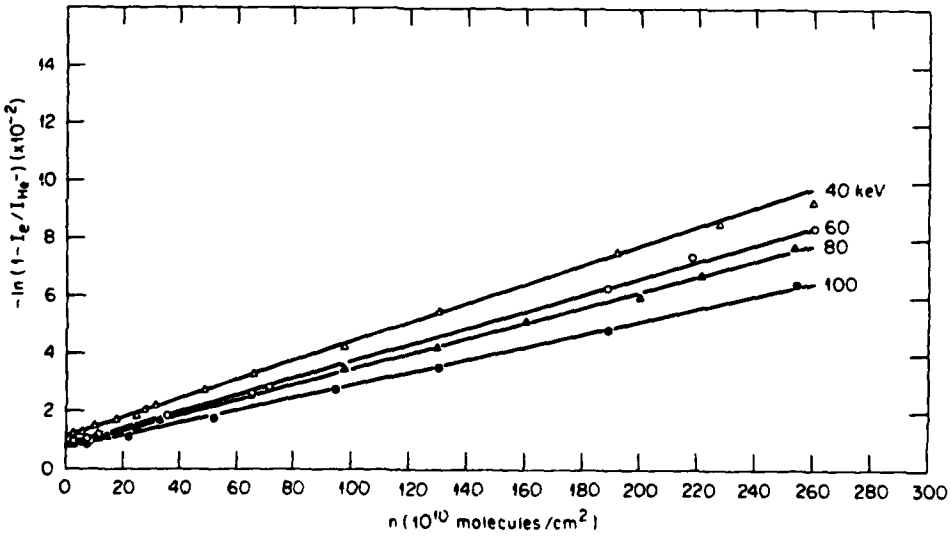

FIG. 2. Dependence of the collisionally induced and autodetaching electron signal vs N_2 molecular density for several ion energies.

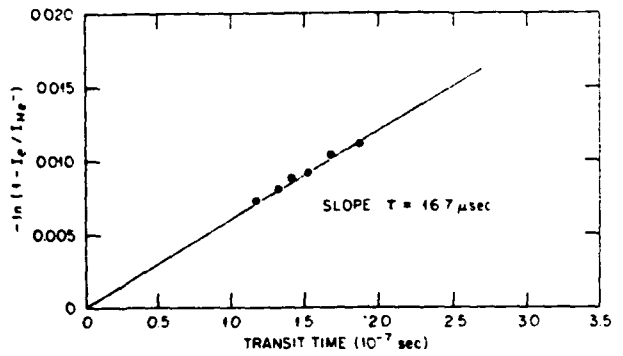

FIG. 3. Dependence of the autodetachment electron signal vs ion-beam transit time.

time of the He$^-$ states present in the ion beam. The lifetime agrees reasonably well with the weighted averages of the short-lived components of the ($^4P^o$)He$^-$ state obtained by Novick and Weinflash[5] when consideration is given to the quoted limits of errors associated with the respective lifetime values. However, the experimental arrangement and ion energies utilized in the present investigations discriminate against long-lived components relative to short-lived components. For example, if we assume the state to be that of the ($^4P^o_J$)He$^-$, in the absence of magnetic fields, the $J=\frac{5}{2}$ components would contribute less than 5% of the collected autodetachment signal at an ion energy of 20 keV—the lowest ion energy utilized in the experiment. As noted, the uncertainties associated with a least-squares fit to the semilogarithmic signal versus ion-beam transit-time data shown in Fig. 3 are small. However, we expect determinant (systematic) errors to dominate and estimate them to be approximately ±15%, which leads to an uncertainty in τ of ±2.5 μsec. The data were collected during two sets of experimental measurements separated by a time interval of a few months.

III. ELECTRON ENERGY MEASUREMENTS

A. Experimental techniques

As a more definitive test of the presence of long-lived ($^2P^e$)He$^-$ states, the energies of autodetaching electrons from the He$^-$-ion beam produced in the Ca charge-exchange medium were measured by replacing the parallel-plate arrangement with a high-resolution double-focusing 180° spherical-sector electrostatic energy analyzer with typical resolution between 0.2% and 0.5% (see the inset of Fig. 1). Although electrostatic energy analysis has been utilized for a number of years in fundamental research, it has not been used previously for measuring autodetachment electron energies from He$^-$ or other long-lived metastable-state negative ions. For reasons previously discussed, the Ca cell again was operated at power levels ~10% of those required for equilibrium production and loss He$^-$ formation. The momentum-analyzed He$^-$ beam was monitored in a biased and shielded Faraday cup located in the measurement chamber with a small hole located in the base of the cup which served as a collimating aperture preceding a 1.5-cm-long gas cell. The purpose of the cell was to provide a high-pressure region for collisionally stripping electrons from the He$^-$ beam when a small amount of gas was introduced in the cell. Electrons, moving collinearly with the ion beam which were stripped from the ion beam due to collisions in the gas cell or ejected in the forward direction by autodetachment in the 5-cm-long drift space following the cell were measured with the analysis system previously described. The beam collimation system used in the measurements permitted angular acceptances α of ±1.5° with respect to the beam axis. The ion beam, after transit through the entrance aperture of the analyzer, was allowed to pass through a high-transmission gridded aperture located in the rear of the analyzer and into a shielded and biased Faraday cup where it was monitored. The electron spectrometer was operated in a fixed pass energy mode thus requiring the acceleration of electrons moving at energies less than and deceleration of those moving greater than the pass energy of the analyzer. This was accomplished by linearly varying the voltage across a single-gap electrode system at the entrance of the analyzer.

The experimental chamber was equipped with three sets of mutually perpendicular Helmholtz coils which were used to nullify the earth and stray magnetic fields in the vicinity of the electron spectrometer which could, otherwise, distort the energy spectrum. Electron spectra were taken with a standard x-y recorder with abscissa and ordinate directions, respectively, driven by signals from a precision ramp voltage generator and a conventional channel electron-multiplier circuit. A typical count rate at the peak of the autodetachment signal was $\sim 10^4$ electrons/sec.

The energies of autodetaching electrons were determined by measuring the energy difference between the auto-detachment and collisional-detachment electron peaks—the latter of which could be enhanced by introducing a small amount of gas in the cell preceding the analysis system. However, this procedure was not required because of the presence of a strong pressure-independent electron peak which appeared at a position corresponding to the velocity of the He$^-$-ion beam. The origin of this low-energy peak is, in part, attributable to electron photodetachment of He$^-$ by ambient blackbody radiation and is currently the subject of independent investigations.

The observed laboratory energy E_{lab} of an electron of energy E_e and mass m_e ejected collinearly with respect to a moving ion of energy E_i and mass M_i is given by the following small-angle kinematical approximation:

$$E_{lab} \cong \frac{m_e}{M_i} E_i \pm 2 \left[\frac{m_e}{M_i} E_e E_i \right]^{1/2} + E_e \,, \tag{1}$$

where the positive and negative signs refer, respectively, to ejection of electrons in the direction of or opposite to the motion of the beam. In the present experimental arrangement, only those electrons moving in the direction of the beam could be detected. The electrons ejected in the backward direction were embedded in a high background due

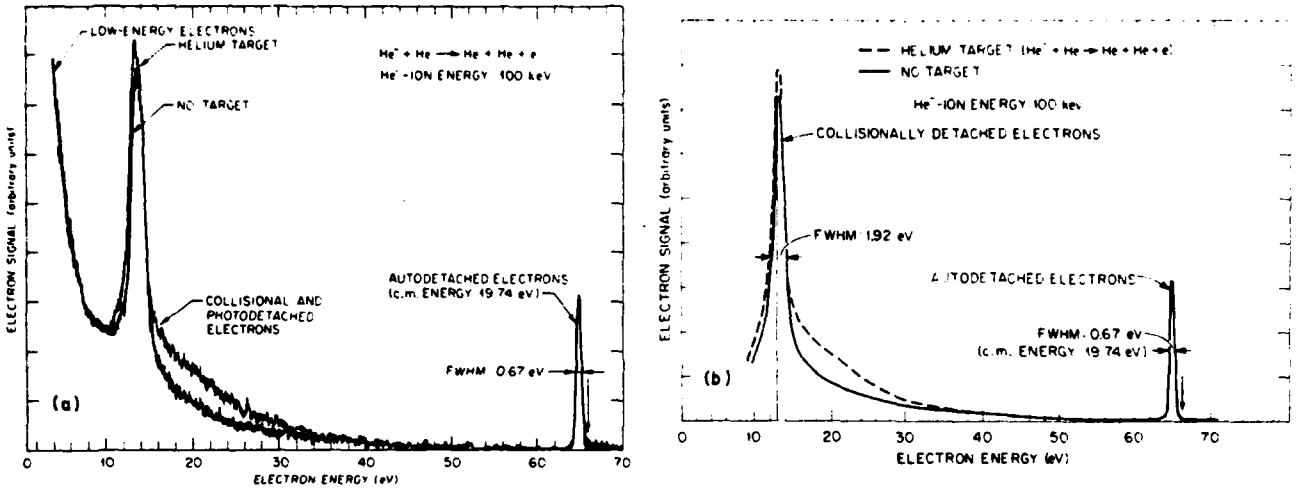

FIG. 4. Collisional and autodetaching electron spectrum from 100-keV He^- ions with and without N_2 stripper gas. Arrow indicates the position in energy where the 20.5-eV autodetaching electron peak from $(^2P^o_j)He^-$ would occur if present: (a) prior to background subtraction and (b) after background subtraction.

to low-energy electrons and, therefore, could not be resolved.

The center-of-mass energy E_e of autodetachment electrons ejected in the forward direction from a moving ion beam of energy E_i can be determined relative to the electrons which move at or near the projectile velocity by use of expression (1). The resulting relation for E_e is given by

$$E_e = \left[\left(\frac{m_e}{M_i} E_i + eX_i - eX_e \right)^{1/2} - \left(\frac{m_e}{M_i} E_i \right)^{1/2} \right]^2, \quad (2)$$

where X_i and X_e are, respectively, the potentials through which the electrons moving at or near the velocity of the ion beam and the autodetaching electrons are accelerated or decelerated upon entrance into the analyzer, and e is the electronic charge. By using this procedure, based on differences, errors due to contact and surface potentials are minimized and the need for precise knowledge of the spectrometer constant and energy-scale calibration is avoided.

B. Results and discussions

Electron energy spectra were determined for a range of ion energies E_i between 20 and 120 keV. Examples of spectra taken at $E_i = 80$ and 100 keV are displayed in Figs. 4 and 5 which are illustrative of more than 20 determinations made over the previously indicated range of ion energies. The data of Fig. 4(a) shown in Fig. 4(b) have been corrected for background. The origin of the low-energy electrons with peak centered at $E_i = 0$ is attributed to secondary electrons generated during collisions of a portion of the He^--ion beam with apertures at the entrance to the analyzer. Figure 5 displays the region surrounding and including the autodetachment electron spectrum taken on a sensitive channel electron-multiplier scale. Among the factors which contribute to the observed widths of the electron energy spectra indicated in Figs. 4 and 5 are the physical processes involved in their formation, the spectrometer resolution function, and the kinematical broadening factor. The resolution function is dependent on spectrometer aperture sizes and the accepted electron-beam angular divergence. In turn, the resultant angular divergence is affected in an energy-dependent way by the lensing action introduced by the acceleration or deceleration of electric fields at the entrance to the analyzer.

Assuming equal production rates of the $^2P^e$ and $^4P^o$ states in the source, the experimental arrangement for measuring autodetachment electron energy levels should

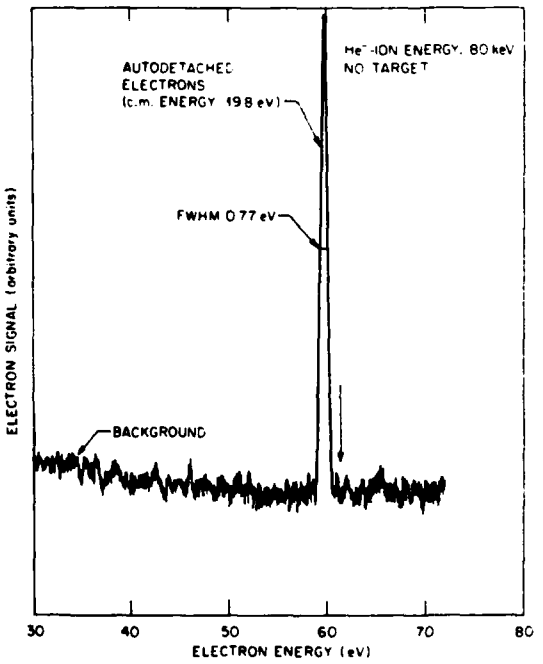

FIG. 5. Display of the region surrounding and including the autodetaching electron peak from 80-keV He^-. Arrow indicates the position in energy where the 20.5-eV autodetaching electron peak from $(^2P^o_j)He^-$ would occur if present.

have permitted detection of $^2P^e$-state populations with lifetimes as short as 0.5 μsec at the highest ion energy (120 keV) utilized in the measurements. No reproducible evidence of additional well-resolved peaks or distortion of the prominent autodetachment peak was observed which would suggest the presence of populations of other decaying states within a factor of ~ 50 in intensity of the principal peak. The positions of the arrows shown in all spectral-data figures indicate the energies at which the 20.5-eV autodetachment peak would have occurred. Again, the presence of the peak would act as a signature of the $(1s2p^2)^2P^e$ He^- state—assuming, of course, the existence of the state and that it lies above and radiatively decays to the promptly autodetaching $(1s2s2p)^2P^o$ He^- state.

The autodetaching electron energy level determined from more than 20 measurements is compared with other experimental and theoretical investigations of the $(^4P^o_j)$ states of He^- in Table III. The agreement between the present energy-level measurements and those obtained by others for the $(^4P^o_j)He^-$ state is taken as conclusive evidence that the predominate long-lived He^- state generated in Ca vapor is the $^4P^o_j$ even though the experimental conditions utilized were compatible with formation of $(^2P^e)He^-$ as well.

TABLE III Comparison of previous and present energy levels for metastably bound He^-.

$E[(1s2s)^3S]-E[(1s2s2p)^4P]$ (eV)	$E[(1s2s2p)^4P]$ (eV)	Reference
≥ 0.075	19.744	1 (Theor.)
≥ 0.033	19.786	6 (Theor.)
0.067	19.752	7 (Theor.)
0.0774 ± 0.0003	19.7417	8 (Theor.)
0.080 ± 0.002	19.79	11 (Expt.)
0.076 ± 0.002	19.74	10 (Expt.)
0.056	19.76	Present

IV. CONCLUSIONS

Experimental investigations of the lifetime and energy levels of metastably bound He^- states formed in charge-exchange interactions with Ca vapor have been made under conditions compatible with the formation of $(^2P^e)He^-$—a state previously conjectured to exist based on the interpretation of experimental data[26–28] as well as the theoretical calculations of Safronova and Senashenka.[30] In contradistinction, the recent configuration-interaction calculations of Bunge and Bunge[8] show the state to be unbound. The present experimental results are supportive of the latter theoretical findings within the limitations imposed by the experimental arrangement on the lifetime of states which could be observed. Both lifetime and energy-level values determined in the present experiment agree closely with those measured or calculated previously for the $(^4P^o_j)He^-$ states. No other structures were observed over an energy range encompassing the anticipated autodetaching energy of the $(^2P^o_j)He^-$ state.

ACKNOWLEDGMENT

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